Recent Developments in Membrane Filtration for Wastewater Treatment

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Abstract Freshwater resources are limited and are becoming increasingly polluted due to the rapid urbanization and industrialization. Water pollution is a preeminent pervasive problem affecting the lives of more than 785 millions people globally, both in terms of quality as well as scarcity. Due to boom in industrialization, several toxins and chemicals such as inorganic particles, harmful hydrocarbon, organic matter, and heavy metals etc. are discharged into freshwater bodies thereby making it unsuitable for domestic and drinking purposes. Therefore, it is imperative to design and perform wastewater treatment processes for the production of freshwater. Various technologies have been explored for this purpose including electrochemical oxidation, advanced oxidation process, advanced biological treatment employing algae, bacteria and fungi and membrane-based filtration techniques. Among these, membrane technology is the most suitable strategy applied for wastewater treatment and has gained considerable attention due to its exciting features such as high separation performance, smaller footprint area, cost-effectiveness, low energy requirement, convenience in operation and high efficiency. In this chapter, we will initially discuss membrane technologies applied for the treatment of wastewater. Then, we will describe various types of synthetic membranes, membrane processes and membrane modules being used in wastewater purification. Afterward, an insight into the membrane operation that includes membrane performance, membrane selectivity, separation mechanism, concentration polarization and membrane fouling will be discussed. Finally, different membrane cleaning processes such as physical, chemical, biological and physicochemical cleaning methods will be discussed.

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1 Introduction

Freshwater is indispensable for life, food, security, public health and energy management on earth. With the increasing urbanization and boom in the human population, the demand for freshwater has excessively increased. The growing scarcity of freshwater bodies is not only alarming for the survival of human and aquatic lives but also increasing global pollution abruptly. According to a recent report, more than 785 million people are facing water scarcity, and the number is increasing gradually (Ilahi et al. 2021). Therefore, conversing the freshwater and purifying wastewater technology is the research hotspot that should be carefully managed and technically fixed in recent times.

With the rapid industrialization and increased urbanization, a large number of pollutants are directly discharged into freshwater bodies, thus making it inappropriate for drinking and domestic purposes (Mahto et al. 2021). These pollutants include harmful hydrocarbon, organic matter, inorganic particles (sand, grift, rubber residue, ceramics), heavy metals, pharmaceuticals and personal care products, pesticides and other related chemical components (Issakhov et al. 2021). Where the organic pollutants, i.e. dyes, textile and food waste, plant material, and paper fibers are among the key contributor to polluting water. These pollutants adversely affect the water quality and risk chemical oxygen demand, alter the chemical composition of water and imparts deep coloration, which ultimately increases the toxicity and decreases biodegradability of freshwater (Werber et al. 2016; Nqombolo et al. 2018; Karri et al. 2021; Dehghani et al. 2021).

To conserve the wastewater, various purification methods including advanced oxidation process, electrochemical oxidation, advanced biological treatment utilizing bacteria, algae and fungi, and membrane filtration have been used. Among these, membrane filtration-based technologies have received considerable attention attributed to their intriguing features such as smaller footprint area, high separation performance, low energy requirement, cost-effectiveness and convenience in operation (Ong et al. 2017; Noamani et al. 2019).

2 Membrane Technology

Membrane technology is a widely adopted separation and purification technique (Cui et al. 2010a). As membranes vary greatly in structure, properties and usage, an all-inclusive description that can relate all of its properties is challenging. The membrane can be defined as a synthetic film that separates two phases by restricting or allowing the passage of various components through it in a selective fashion (Singh and Hankins 2016). The movement of molecules across the membranes is the result of convective movement or diffusion of molecules. Classification of different membrane processes is primarily characterized by different propelling forces, including pressure, concentration, temperature and membrane pore size. At the same time, its thickness ranges from several centimetres to less than 100 nm and the average pore diameter range from several micrometres to less than 0.1 nm. Membranes are largely governed by different force gradients such as osmotic pressure, concentration, applied pressure, electrical and thermal or the combination of these driving forces (Khan et al. 2021a; Lau et al. 2020). For instance, a membrane sheath can be nonporous or porous, anisotropic or isotropic and electrically charged or neutral (Strathmann 1986).

The choice of the membrane type and the process is determined by numerous factors, which include feed mixture, degree of separation projected and feed volume needed to be processed.

3 Types of Membranes

Synthetic membranes are of two types, i.e. solid or liquid. Based on the morphology, solid membranes are categorized as isotropic or anisotropic (Purkait et al. 2018). Isotropic membranes are also sometimes called symmetric membranes whose composition and physical structure are uniform all over the membrane. Isotropic membranes are further classified as microporous, nonporous, and electrically charged membranes. Isotropic microporous membranes are rigid, having highly voided structures with interconnected and randomly distributed pores that have higher permeation fluxes (Sagle and Freeman 2004). Nonporous isotropic membranes comprise a dense sheath where permeate diffuses under the effect of different propelling forces such as concentration, electric potential and pressure gradient. The separation of different mixture components and their relative transport across a membrane is reliant on their solubility and diffusibility in the membrane. These membranes have comparatively lower permeation fluxes than porous membranes, due to which their applications are limited (Obotey Ezugbe and Rathilal 2020). Electrically charged isotropic membranes are either porous or dense but are exquisitely microporous in most cases (Purkait et al. 2018). These are cationic exchange or anionic exchange membranes, whose pore walls contain fixed negative or positive charged ions, respectively. These membranes are permeable to oppositely charged ions but repel similarly charged ions (Jun et al. 2020; Jua et al. 2020). The ion charge and concentration in the solution drive the separation process. Electrodialysis reversal is the most common application of electrically charged membranes (Xu 2005). Conversely, anisotropic membranes can be asymmetric or composite membranes, which are irregular all through the membrane area and consist of multiple layers having diverse compositions and structures. These membranes are composed of a very fine selective film, which is assisted by a highly permeable and dense sheet and is specifically useful in reverse osmosis applications (Mallevialle et al. 1996).

A liquid membrane consists of a liquid phase that exists as a supported or rather unsupported form that works as a membrane fence to separate two distinct phases of a solution (Hansen et al. 2021). The supported form of the liquid membrane comprises a microporous assembly that is occupied by a liquid membrane phase. The microporous assembly and the liquid-filled pores provide the much-needed mechanical strength and selective separation barrier, respectively. Higher porosity and smaller pore size are the key contributors to maintaining the liquid phase under hydrostatic pressure. These membranes are extensively applied both analytically and industrially for preconcentration, purification and treatment of wastewater (Parhi 2013). Conversely, an unsupported liquid membrane or emulsion liquid membrane is an emulsion-type mixture that consists of a thin liquid film and is equilibrated by the action of a surfactant. These membranes have tremendous potential for the treatment of wastewater containing heavy metals and hydrocarbons owing to their simple operation, removal and stripping in a single stage, high efficiency, superior interfacial area, and choice of continuous operation (Kumar et al. 2019; Baker 2012).

Based on membrane material, synthetic membranes are categorized as organic, inorganic, and hybrid or mixed matrix membranes (Khan et al. 2021b). Polymeric or organic membranes are synthesized from synthetic organic polymers, i.e. polytetrafluoroethylene, polyethylene, cellulose acetate and polypropylene, etc. Mostly, these polymers are utilized for the synthesis of polymeric membranes for processes that are driven by a pressure gradient, which includes ultrafiltration, microfiltration, nanofiltration, and reverse osmosis (Aliyu et al. 2018).

Inorganic membranes are synthesized from silica, ceramics, zeolites or metals. These membranes are stable in an intensive thermal and chemical environment and are extensively used for industrial applications such as ultrafiltration, microfiltration and hydrogen separation (Mallada and Menéndez 2008).

Nowadays, mixed matrix membranes or hybrid membranes have gained considerable attention. Mixed matrix membranes are considered next-generation hybrid membranes materials that combine the inherent properties of both the polymer and the fillers. The hybrid membrane overcomes limitations of the polymeric and inorganic membranes while synergizing and utilizing properties of both, i.e., easy and viable processability of the polymers and enhanced selectivity of the inorganic fillers. The polymeric material is utilized as a continuous phase in which various fillers are dispersed (Qadir et al. 2017).

4 Membranes Processes

The movement of the material through the membrane is the result of various driving forces, including pressure difference, concentration, electrical potential and temperature gradient or a combination of these processes (Jhaveri and Murthy 2016).

4.1 Pressure Difference-Based Membrane Processes

Pressure difference-based membrane processes have been the most extensively applied practices for the purification of wastewater. These practices are applied from pretreatment to posttreatment of wastewater. Two factors are responsible for

Table 1 Main characteristics of pressure difference-based membrane processes

Process	Pore size (μm)	Pressure range (bar)	MWCT* (kilo Dalton)	Average permeability (L/m^2 h bar)	Solutions retained
Microfiltration	0.05–10	1–2	100–500	500	Bacteria, fat, oil, organics, colloids, microparticles
Ultrafiltration	0.001–0.05	2–5	20–150	150	Proteins, oils, pigments, organics, microplastics
Nanofiltration	<0.002	5–15	2–20	10–20	Pigments, divalent anions, cations, sulfates, lactose, sucrose, sodium chloride
Reverse osmosis	~0.0006	15–100	0.2–2	5–10	All impurities containing monovalent ions

* MCWO = molecular weight cut off

separation processes, i.e. the transmembrane pressure and the decreasing membrane pore size (Chollom 2014). These processes are grouped into four types based on transmembrane pressure and membrane pore size, i.e. microfiltration, nanofiltration, ultrafiltration, and reverse osmosis. Table 1. provides the principal characteristics of these processes.

4.1.1 Microfiltration

In microfiltration, the pressure gradient drives the membrane separation process. The pore size of the microfiltration membrane and the pressure lie in the range from 0.05–10 μm and from 1–2 bar, respectively. Microfiltration is widely used for the elimination of microbes, particulates, and turbidity and is often used as a preprocessing step to different pressure-related membrane processes (membrane distillation, ultrafiltration and reverse osmosis) (Singh 2006). Urban wastewater and drinking water production are two main applications of MF. Also, it is used for the cleansing of wastewater from the oil industry, heavy metal wastewater and paint industry (Obotey Ezugbe and Rathilal 2020).

4.1.2 Ultrafiltration

Ultrafiltration is a pressure-gradient centered membrane operation that has a pore size ranging from 0.001 to 0.05 μm and a pressure range from 2–5 bar. Ultrafiltration eliminates macromolecular solutes, viruses, suspension, fine colloids, organic material, and other contaminations from water. It is used in the treatment of industrial wastewater (textile, oil, and pulp industry), electrophoresis coating wastewater, and wastewater having heavy metals, enzymes, and starch (Peters 2010).

Microfiltration and ultrafiltration membranes are usually made by phase inversion method and have comparatively wider pore distribution, due to which it offers certain disadvantages, including higher sensitivity to fouling and are susceptibility to pore blocking. A larger pore also lets certain species pass, which is to be retained (Obotey Ezugbe and Rathilal 2020).

4.1.3 Nanofiltration (NF)

NF is an advanced molecular level membrane process prompted by the difference in transmembrane pressure through the membrane. NF falls in the middle of ultrafiltration and reverses osmosis (RO). It can also be termed as loose RO or low-pressure RO. It separates molecules using a nano-porous permselective membrane having a pore size of less than 0.002 μm corresponding to molecular weight cut off ranging from 200–2000 g/mol and involving the pressure of 5–15 bar, nearly at an ambient operational temperature. These membranes are capable of the exclusion of inorganic salts and minute organic molecules. The distinctive features of NF membranes are a lower and higher rejection of monovalent and divalent ions, respectively, and greater fluxes in comparison to RO membranes. These features qualify NF membranes for widespread applications, including food engineering, pharmaceutical, biotechnology, and especially for wastewater treatment, i.e., natural organic removal, water softening, pretreatment to remove scale former in thermal desalination and plays the same role prior to reverse osmosis to increase the saltwater RO (SWRO) recovery (Fane et al. 2015).

4.1.4 RO

RO is the most sophisticated pressure-related membrane purification technology (Joo and Tansel 2015). The membrane pore size is approximately in the range of 0.0006 μm, thus preventing all the dissolved solids, sediments, colloidal particles and microorganisms. In osmosis, water usually moves towards the salty concentrated solution, however, in RO the water from the concentrated side infiltrates through the semipermeable membrane in the presence of an external applied hydraulic pressure which is about 15–100 bar in range (Macedonio and Drioli 2010). RO eliminates particles in the molecular weight cut-off range greater than 50 Daltons which include suspended solids, dissolved salts and matter, colloids, organic matter, bacteria, viruses, trihalomethanes and volatile organic compounds (Abd El-Salam and Caballero 2003). RO has widespread applications including brackish water desalination and treatment of emissions from chemical, pharmaceutical, textile and other industrial processes. The major disadvantages of RO during wastewater treatment are membrane damage due to the presence of strong bases, acids, free chlorine, and membrane fouling because of concentration polarization by inorganic, organic, metal oxides, and biological matter. RO membrane eliminates all the contaminants in a solitary step and offers an ideal and economical water cleansing process (Bartels et al. 2005).

4.2 Concentration Gradient Based Membrane Processes

4.2.1 Forward Osmosis (FO)

FO is an osmosis driven membrane-based process wherein water molecules are forced to permeate through a semipermeable membrane from a feed solution (FS) to a draw solution (DS) as a consequence of concentration difference, which offers the much-needed difference in osmotic pressure as shown in Fig. 1a. Unlike, RO which exploits the hydraulic pressure differential, FO employs the osmotic pressure differential ($\Delta\pi$) as a propelling force across the membrane. Subsequently, in FO, the diluted feed or introductory solution becomes concentrated while the highly concentrated draw solution is diluted as a result of water transport from FS to DS. FO continues until potential chemical equilibrium is established (Suwaileh et al. 2018). Except for applications, where the drawn water becomes part of the draw solution, a recovery unit is always needed that continuously extracts pure water and restores the concentration of the draw solution. In the course of FO process, the draw solution generates the driving force on the membrane permeate side. The foremost criteria for selecting a draw solution is that its osmotic pressure must be greater than the feed solution. FO is surely the most proficient membrane-based purification process in the treatment of industrial wastewater (Haupt and Lerch 2018).

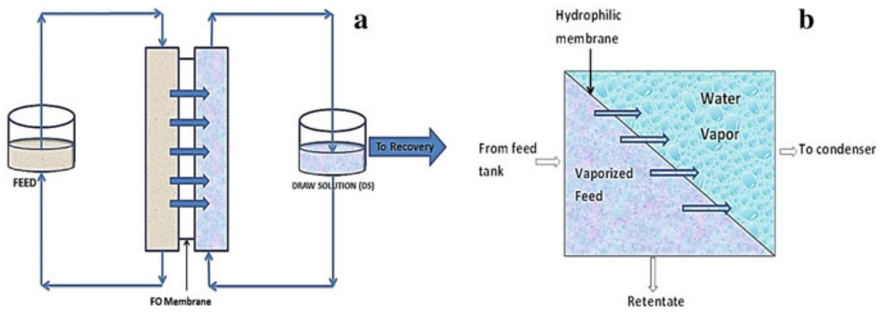

Fig. 1 Schematic illustration of forwarding osmosis **a** and pervaporation **b** (Obotey Ezugbe and Rathilal 2020)

4.2.2 Pervaporation

Pervaporation is a membrane-centered separation practice that couples evaporation with membrane permeation for the preferential purification of liquid mixtures, as shown in Fig. 1b. The feed stream, which usually contains two or more components, interacts with one side of the membrane while the membrane permeates side is conditioned with vacuum or sweeping gas. Different components in the liquid stream are sorbed onto the membrane upstream to generate vapor permeate downstream, and a liquid reject. The feed stream is processed at a temperature and pressure of about 100 °C and 1 atm respectively, however, maintaining a vaccum at the membrane permeate side. The infiltrate vaporizes as the infiltrating components pass through the membrane. The most commonly used membrane for PV is hydrophilic asymmetric composite membranes, including polyvinyl alcohol/polysulfone or PVA/polyacrylonitrile composite membranes (Kesting and Fritzsche 1993). Also, thin layer PDMS/PAN membranes were found effective for the separation of polar and nonpolar compounds. Frequently used membrane modules are spiral wound and flat sheet modules. PV is employed for the treatment of wastewater for micro-irrigation, removal of organic solvents from aqueous solution, removal of alcohols from fermentation broth and also for recovering isopropanol from water with 99% purity (Singh 2014).

The structure and chemical nature of the pervaporation membrane has a consequential function in the separation process. Therefore, these membranes are specifically devised to achieve maximum affinities for intended applications (Basile et al. 2015). Other key features that affect the pervaporation process include partial pressure, feed concentration, temperature, and feed flow rate (Gongping et al. 2011). Unlike conventional separation processes, pervaporation is environmentally friendly and energy-saving technology. However, due to sensitive operational parameters, its industrial utilization is still not achieved (Huang and Meagher 2001).

4.3 Electric Potential Gradient-Based Membrane Processes

4.3.1 Electrodialysis and Electrodialysis Reversal

In electrodialysis, ions are directed across an electrically charged membrane due to differences in electric potential. The electrodialysis cell consists of alternating cationic and anionic exchange membranes that are positioned in between the vicinity of two electrodes, as shown in Fig. 2. The feed solution drifts through each membrane pair in the cell. On the application of an electric potential gradient across the system, cations and anions travel through charged membranes towards their respective electrodes, i.e. cations approaching cathode and anions approaching anode. The cation exchange membrane permits cations to pass through, but anions are rejected, and the anion exchange membrane lets anion flow through but rejects cation permeability.

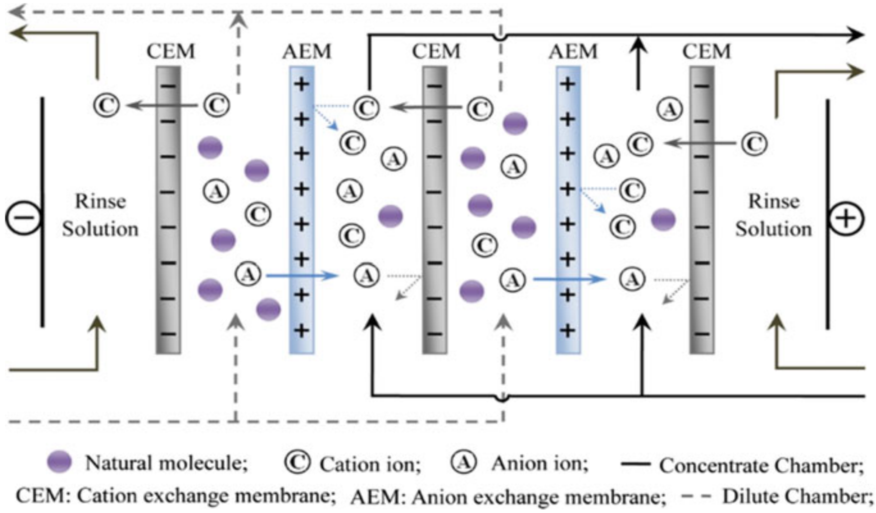

Fig. 2 Schematic representation of ion exchange membranes (Ran et al. 2017)

The arrangement of electrically charged membranes is such that there is an alternation of chambers having greater ion concentration and chambers with a very trivial ion concentration. The high concentrated ion solution is called the concentrate, while the dilute solution almost free of ions is called the dilutate, which is the product of water (Baker and Wilson 2000).

In electrodialysis reversal (EDR), the movement of ions across the membranes is reversed by the periodic reversal of the electrodes. As a result, the concentrated solution becomes dilute, and the dilute solution becomes concentrated, which acts as a self-cleaning feature, thus decreasing fouling and extending membrane life (Obotey Ezugbe and Rathilal 2020).

Electrodialysis and electrodialysis reversal have immense applications in treating wastewater, especially for the elimination of total dissolved solids and ionizing species in water. ED and EDR have several important features, which include a high recovery rate of water, slight pretreatment of feed water and reduced membrane fouling because of process reversal. However, ED is ineffective in the desalination of highly saline water since the energy required for desalination is directly proportional to the removal of ions. Also, it cannot remove unionized compounds and harmful substances such as bacteria and viruses, and hence posttreatment will be required, which elevates the process cost. Moreover, chlorine formation at anode results in corrosion (Obotey Ezugbe and Rathilal 2020; Chao and Liang 2008).

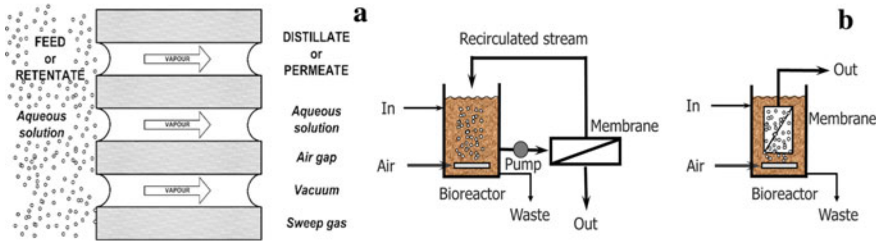

Fig. 3 Schematic representation of membrane distillation **a** (Curcio and Drioli 2005) and Membrane bioreactor **b** (Judd 2008)

4.4 Hybrid Membrane Processes

4.4.1 Membrane Distillation (Temperature Gradient-Based Membrane Processes)

Membrane distillation, a hybrid membrane purification method that integrates membrane technology with thermal distillation, as shown in Fig. 3a (Belessiotis et al. 2016). Two liquids are separated due to differences in partial pressure and temperature across the membrane. The feed is charged at a temperature of up to 100° C while the permeate is cooled down by applying distilled water. The difference in temperature via membrane creates a difference in vapor pressure that drives the H_2O vapor molecules or volatile constituents from hot feed solution through a nonporous hydrophobic membrane. Separation comes about by vaporization followed by vapor passage across the membrane pore opening and membrane network. The difference in vapor pressure is proportional to the water vapor flux. The increase in temperature difference across the membrane causes an exponential increase in the vapor pressure, which accelerates the flux substantially. Salinity in the feed stream reduces the driving force, i.e. vapor pressure, however, this effect is negligible unless salinity is too high (Mulder and Mulder 1996).

MD is an effective process in the purification of highly saline water(seawaters) and rejects brine concentrates of RO as high osmotic pressure cannot affect the process. However, several factors negatively affect the process performance, such as pore wetting, fouling, poor heat transfer, heat loss, low mass transfer due to air entrapment in the membrane pores, and high energy consumption (Gongping et al. 2011).

4.4.2 Membrane Bioreactor (MBR)

In MBRs (Fig. 3b), biological wastewater purification routes such as activated sludge processes are coupled with membrane practices including MF, UF, and NF that are extensively applied in municipal and industrial wastewater processing. MBRs are increasingly being applied in wastewater treatment due to several advantages such as increased pollutant removal, low footprint required, and a reduced amount of sludge yield. In membrane filtration, microbes are entrapped in the biological reactor, which affords superior regulation over the biological reactions and modifiable parameters of microbes in the ventilated chamber. Thus providing an increased accumulation of mixed liquor suspended solids (MLSS) and prolonging solid retention time (SRT) (Stephenson et al. 2000). Generally, MBRs are categorized into three kinds based on operational mechanisms: rejection MBR, diffusive MBR, and extractive MBR. MBR procedures are effectively implemented in the treatment of small-scale industrial wastewater purification plants and large-scale wastewater purification plants. Mostly two types of configuration are used in MBR; (1) side stream MBR and; (2) immersed MBR. The membranes in sidestream MBR are installed externally to a bioreactor which needs a pumping system for transporting biomass for the filtration process and residue back to the reactor from the filtration unit. The advantage of this setup is that cleaning of an externally installed membrane can be done easily. However, side stream MBR has limited application due to higher energy and pressure requirements. In immersed MBR, the membrane module is immersed in a bioreactor wherein effluents are forced across the membrane. However, the sludge is stuck in the membrane. Air is usually supplied for sustaining aerobic settings and cleaning and scrubbing the exterior and surface of the membrane, respectively. Immersed MBR is used more commonly as compared to side stream MBR due to its simple operation and low energy consumption, however, cleaning the membrane module is difficult as it is submerged in the bioreactor. MBR based process has several benefits in comparison to conventional treatment processes that include production of high quality clarified water, smaller footprint, better regulation of solid and hydraulic retention time, and provides a fence to chlorine-resistant pathogens due to effective membrane pore size less than 0.1 μm. Moreover, designing long sludge age MBR can achieve low production of low excess sludge, thus endorsing the enrichment of nitrifying bacteria, which in turn increases the removal of nitrogen (Wen et al. 2010).

5 Membrane Module

The membrane module is how single operation units are designed and engineered into devices and hardware to attain the anticipated separation performance. It is composed of a membrane, feed inlet and outlet points, permeate draw-off points and pressure support structures (Obotey Ezugbe and Rathilal 2020). So far, four kinds of membrane modules are in use in the industry, which is briefly discussed:

i. Tubular modules
ii. Hollow fiber modules
iii. Flat sheet modules
iv. Spiral wound modules

5.1 Tubular Modules

The tubular module is composed of an outer covering called a shell which is tube-like in nature, as demonstrated in Fig. 4a. The tube-shaped shell is composed of a permselective membrane implanted inside poriferous fiberglass or stainless steel. It contains about 30 porous tubes having a diameter ranging from 0.5 to 1.0 cm. The feed to be processed is passed through the tubes by applying pressure, and the infiltrate is concentrated via infiltrating opening on the shell side. Tubular modules have some distinctive features: (1) these are adapted for feed streams with bigger particle sizes because of their large inner diameters. Additionally, their chemical or mechanical cleaning is easy; (2) they usually have a turbulent flow condition with Reynolds number >10,000 and need huge pumping capacity; (3) their surface area to volume ratio is the smallest among all the membranes modules and consequently

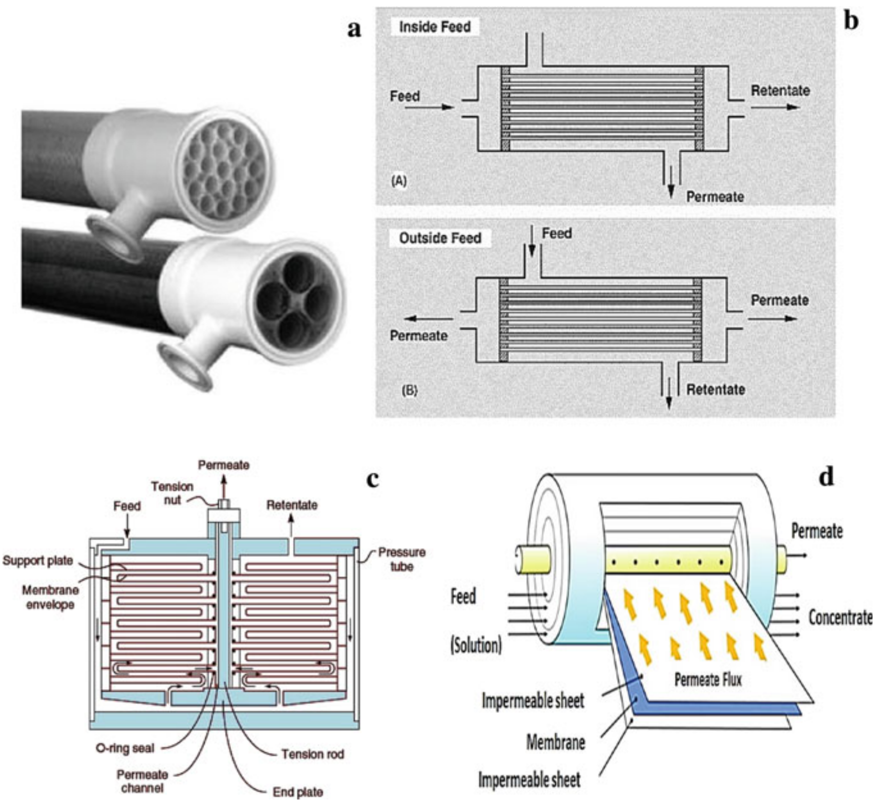

Fig. 4 Schematic representation of tubular **a** (Berk and Berk 2009), hollow fibre **b** (Bruggen et al. 2015; Bruggen et al. 2015; Heidelberg. 2015), flat sheet **c** (Baker 2012) spiral wound modules (Obotey Ezugbe and Rathilal 2020)

have high hold up a volume that necessitate huge flooring space to function (Cui et al. 2010b).

5.2 Hollow Fiber Modules

Hollow fiber modules (Fig. 4b), in principle, are analogous to tubular modules arrangement. As the name indicates, hollow fibers consist of thin tubes, the diameters of which range from 1 nm to capillary size fibers. These fibers, being self-supported, have a high backflushing capacity. It contains about 50–3000 hollow fibers, which are connected to porous end plates, and the intact package is inserted into a jacket or vessel. They may have outside in or inside-out flow direction (Cui et al. 2010b). Their key distinctive features, which are different from tubular modules, are: (1) mostly they have laminar flow characteristics which correspond to a Reynolds number of 500–3000. Also, their pressure domain is low, around 2.5 bar at maximum; (2) they are very economical concerning energy consumption because of low pressure and crossflow rate; (3) among all the membrane modules, hollow fibers modules possess the highest ratio of surface area to volume and have low holdup volume; (4) due to their self-sustaining characteristic, they have improved backflushing capability and are cleaned easily; (5) one characteristic shortcoming of hollow fiber module is their liability to get clogged by large size particles during the inside-out operational mode, thus it requires pretreatment to decrease the particle size to 100 μm (Singh and Hankins 2016).

5.3 Flat Sheet Module

The flat sheet module (Fig. 4c) is composed of an upper selective horizontal membrane sheet and bottom flat plate. A mesh-like material in between the upper sheet and the bottom plate is placed for removal of permeate, and across the flat plate, an additional membrane sheet and mesh-like material is positioned in the mirror, which forms a sandwich resembling module. The channel gaps and length in these modules range from 0.5 to 10 mm and 10 t0 60 cm, respectively. Their Reynolds number corresponds to laminar flow characteristics, however, screening the feed channel results in better mixing. The pretreatment required to decrease the particle size up to 120 μm is recommended. The advantage of flat sheet modules is that they can be cleaned easily by taking out the membrane, and thus fouling can be controlled, however, they are less efficient due to their low packing density. In terms of energy requirement, cost, and packing density, these modules lie in between spiral wounds and tubular modules (Zirehpour and Rahimpour 2016).

5.4 Spiral Wound Modules

Spiral wound membrane modules (Fig. 4d) show resemblance to flat sheet membrane modules. These membrane modules are composed of large membranes which are wrapped around a central perforated collection tube. A mesh-like permeable plate or is inserted between two membrane sheets, the active membrane sides of which are facing away with the supporting side facing the feed distributor directly. The feed stream runs aligned to the central collection pipe wherein the permeate falls perpendicular to the feed stream flowing through the permeate spacer. On an industrial level, a tubular pressure vessel contains around six spiral wound modules which are 40 inches long having a diameter of around 8 inches. Spiral wound modules have several important features: (1) the modules flow characteristics is turbulent due to the existence of feed spacers; (2) these modules have relatively high pressure drop due to the surplus drag caused by the presence of feed spacers; (3) the surface to volume ratio is relatively high and are economical among all the membrane modules; (4) the feed stream may contain suspended particles which can block the mesh-like spacer which in turn can partly block the flow of feed channel. Hence pretreatment is required to remove the suspended particles as spiral wound modules require comparatively clear feed having a lesser amount of suspended particles (Cui et al. 2010a).

6 Operation of Membrane

6.1 Membrane Performance

The performance of a membrane depends on the permeate flow and the retainment of dispersed, suspended and dissolved solids by the membrane. Transport characteristics of different solute materials through a membrane rely on two factors, i.e. permeability of the membrane and driving force. Usually, the main driving force is a gradient in concentration, pressure and electric potential past the membrane. Membrane transport performance can also be altered by flow factors like solvent longitudinal convention, axial solute diffusion, and flow prompted solute particle drag on the surface of the membrane. The potential difference past the membrane regulates the amplitude of the driving forces while the transfer of mass from feed to infiltrate side of a solution is controlled by a membrane. The lack of external forces turns the potential difference into zero across the membrane, which develops equilibrium in the system. Membrane processes are not equilibrium processes like evaporation, distillation and crystallization but are kinetic processes, and under constant driving force and steady-state conditions, there is a constant flux through the membrane (Zirehpour and Rahimpour 2016). The correlation between flux and driving force is:

$$J = K \times X \tag{1}$$

where J represents flux or permeate flow, K demonstrates proportionality constant, and X shows the potential gradient/membrane thickness, respectively (Singh 2014).

Numerous semiempirical models like Ohm's law, Fick's and Hagen Poisseuille's have been utilized to describe the mass transport across the membrane. For processes that are driven by the pressure difference (RO, UF, NF and MF), the flux correlation follows as:

$$J = K \times \Delta P/t \quad (2)$$

where J, K, ΔP and t corresponds to transmembrane flux, permeability constant, pressure difference and membrane thickness, respectively (Zirehpour and Rahimpour 2016).

The efficacy of a liquid separation practice can be assessed from the conversion or recovery of the process, which is the combination of total membrane area and flux. The conversion percentage is the percent conversion of feed into the product stream. % recovery is given by

$$R(\%) = \frac{\text{Permeate flow}}{\text{Feed flow}} \times 100$$

6.2 Membrane Selectivity

Membrane material physical and chemical nature is responsible for the membrane separation. The difference in various features of different substances, including shape, aperture, chemical, and electrical charge, are responsible for the separation across the membrane. In porous membranes, the particles are fairly small to permeate through membrane apertures by convective flow, while in nonporous membranes, transport occurs by sorption or diffusion through the membrane (Singh 2014). The selectivity of the membrane is quantifiable vis-à-vis rejection. The rejection coefficient (R) is a dependable criterion for estimating the membrane separation performance of a process

$$\%\text{Rejection (R)} = \frac{\text{feed} \div \text{bulk solute concentration} - \text{product solute concentration}}{\text{feed} \div \text{bulk solute concentration}} \times 100$$

The value of R = 100 for an ideally selective membrane. Membrane selectivity of some routes, i.e. MF and UF, are well explained by the retention coefficient because the solute transport across the membrane is affected by concentration polarization or gel layer formation. % retention is given by

$$\%\text{Retention} = \frac{\text{solute concentration of membrance surface} - \text{permeate solute concentration}}{\text{solute concentration of membrance surface}} \times 100$$

The tradeoff between membrane permeability and selectivity affects membrane performance. Membrane selectivity is related to the permeation of different components through the membrane, i.e. ($\alpha = A/B$), where A represents the permeability coefficient of water and B represents the permeability coefficient of solute (pollute) (Singh 2006).

6.3 Mechanism of Separation Through the Membrane

Membrane separation underlines the ability of membranes to regulate the infiltration of various components or species. Generally, membrane separation mechanisms are of two kinds, i.e., molecular filtration and solution diffusion. In microporous membranes, molecular filtration drives the separation process while dense membranes separation follows the solution diffusion model where the mobility and diffusion of different species in the membrane drive the separation process (Zirehpour and Rahimpour 2016).

In molecular sieving membranes, the separation of feed into different components is brought about by pressure-driven drift through fixed-sized small pores. Different components are separated based on the difference in their sizes. Conversely, the membrane material of solution diffusion membranes is composed of a dense layer of polymer that contains no pores or apertures. The permeants first dissolve in the membrane material and then diffuse across the membrane due to concentration gradient.

The difference in solubility and diffusion rates is responsible for the separation of various components in these membranes. The difference in mechanism of solution diffusion and molecular sieve membranes is due to variance in size and lifespan of membrane pores.

6.4 Concentration Polarization (CP)

CP is actually the growth of the rejected components on or close to the surface of the membrane. It is shared by all the membrane separation processes in which a layer of solute particle mounts up on the membrane periphery as the feed infiltrates through the membrane. During all membrane processes, the passage of feed components towards the surface of the membrane is governed by convection which upsurges as the infiltration across the membrane accelerates. The less permeable components hold back because of membrane selectivity and are conveyed back to the bulk feed at a steady rate (Zirehpour and Rahimpour 2016). The modeling of the membrane module is complicated by CP due to the dissimilarity that exists amidst the wall concentration of solute and the concentration of the bulk feed. The wall concentration of solute is difficult to determine, and a boundary layer film model is applied to define this singularity. The convective drift of solute particles to the membrane periphery and the infiltration of solute particles through the membrane deduct are equal under steady-state conditions with solute particles diffusion back to the bulk feed solution.

$$\mathrm{J.C} = \mathrm{J.C_p} - \mathrm{D_{ij}}\frac{dC}{dz} \tag{3}$$

Equation (3) integration having boundary conditions, i.e. $z = 0$, $C = C_m$, $z_= l_{bl}$, and $C_= C_b$ will give the following equation

$$\mathrm{J} = \left(\frac{D_{ij}}{l_{bl}}\right)\mathrm{In}\frac{C_m - C_p}{C_b - C_p} \tag{4}$$

Equation (4) that develops the film theory model can be applied to find the solute concentration close to the membrane periphery.

Readjusting Eq. 4 will give:

$$\mathrm{C_m} = \left(\mathrm{C_b} - \mathrm{C_p}\right)\exp\left(\frac{J.l_{bl}}{D_{ij}}\right) \tag{6}$$

The solute concentration close to the periphery of the membrane is very crucial for different models of membrane transport. The ratio D_{ij}/l_b can be described as mass transport coefficient and could be found from conventional chemical engineering, i.e. Reynolds number (Re), Sherwood number (Sh), Schmidt number (Sc) and Peclet number (Pe). The permeate concentration and bulk concentration could be obtained by employing analytical instruments.

The membrane filtration unit performance may be adversely affected by CP, which include a reduction in rejection of undesirable solute and water flux, precipitation caused by increased surface concentration beyond the solubility limit, altering the separation characteristics of the membrane, and increase in colloidal and particulate matter that leads to fouling which blocks the membrane surface. Therefore, designing appropriate membrane modules and suitable operating conditions are crucial to anticipate and inhibit the influence of concentration polarization (Ang et al. 2015).

6.5 Membrane Fouling

Membrane fouling can be described as a phenomenon in which dissolved or suspended particles, microbes and organic materials deposit on the periphery of the membrane material or the internal pores of the membrane material, reducing the performance of the membrane. Membrane fouling is categorized into two groups, i.e. reversible or irreversible fouling which depends on the deposition of rejected solutes on the membrane periphery of internal pores of the membrane, respectively (Speth et al. 1998).

Fouling phenomena affects the functional membrane area causing a flux drop below the theoretical capability of the membrane. Subsequently, higher pressure is required for the passage of permeate through the membrane. Membrane fouling adversely affects the overall membrane performance. These include reduction in the functional membrane area, requires more downtime, and increased energy consumption (Kucera 2015).

Different types of membrane fouling depend on and are named on the type of foulant materials such as colloidal, bio, organic, and inorganic fouling (Amy 2008). Colloidal fouling results from the deposition of biological trash, microorganism, lipoproteins, polysaccharides, proteins, clay, silt, manganese oxides, iron and oil, etc. These constituents amass and deposit on the membrane material in due course (Burn and Gray 2016). Biofouling occurs as a consequence of the growth and deposition of organized and coordinated communities of microbes called biofilms on the membrane material. These biofilms contain extracellular polymeric substances (EPS) and microorganisms which results from the adhesion of microorganisms to wet surfaces. These microorganisms nourish from the nutrients in the system and reproduce, which subsequently block the membrane apertures and thus hamper the infiltrate flow (Matin et al. 2011).

Inorganic fouling can be described as the buildup of inorganic salts on the surface of the membrane material. These inorganic salts contain $CaSO_{4,}SiO_2$ and $CaCO_{3,}$ and some other salts (Lisdonk et al. 2000). The less soluble salts in the feed solution precipitate out and stick to the membrane surface during the formation of scales (Shirazi et al. 2010). Organic fouling is the accumulation and deposition of organic compounds onto the membrane surface. These compounds exist in natural organic material, which reduces the permeate passage through the membrane material (Amy 2008).

Several factors are responsible for membrane fouling, such as:

- Feed characteristics, i.e., ionic strength and pH.
- Membrane features such as membrane material and surface properties, distribution of pore size, roughness, hydrophobicity and hydrophilicity.
- Various process factors comprise the transmembrane pressure, time, crossflow velocity and temperature.

All these factors are crucial for determining membrane performance in comparison to membrane fouling (Cui et al. 2010a).

7 Membrane Cleaning: Control of Fouling

Membrane separation processes are principally size segregation-based mechanisms. In general, the excluded solutes cause the membrane fouling and is thus inexorable. Numerous approaches have been suggested for controlling membrane fouling. The significance of these methods relies upon the distinctive characteristic of the nature of the membrane material and the bulk feed solution. These methods comprise turbulent inducers, improvement of membrane material, boundary layer velocity control, and the practice of exterior fields (Jagannadh and Muralidhara 1996). Similarly, pretreatment of feed, rotating membranes, flow handling, and gas sparging and have also been proposed by some researchers (Williams and Wakeman 2000).

Membrane cleaning is aimed at restoring the lost permeability and permeation flux of the membrane as a consequence of membrane fouling. It encompasses the elimination of the amassed material from the membrane periphery to ensure permeability. Cleaning methods for membranes can be classified into physical, biological, chemical, and physicochemical cleaning methods. Additionally, cleaning can be described as ex-situ or in-situ whether the membrane module is washed outside or inside the reactor, respectively (Wang et al. 2014).

7.1 Physical Cleaning

Physical cleaning encompasses the treatment of the membrane module mechanically to eradicate the accumulated material that adheres to the membrane surface. These are of different types, which include periodic backflushing, backwashing, pneumatic cleaning, ultrasonic cleaning, and sponge cleaning (Zhao et al. 2000).

7.1.1 Periodic Backflushing

Periodic backflushing is the backward movement of permeate by applying pressure from permeate side. This backflushing causes the accumulated materials to be detached from the surface of the membrane. The applied pressure required for backwashing should be greater than the filtration pressure (Yigit et al. 2010). Backwashing is the most widely applied industrial method for foulants removal and can successfully redeem the lost flux caused by reversible fouling because of the accumulation of material to the periphery of the membrane material. However, it is ineffective for reversing the irreversible fouling that results from clogging or deposition of suspended or dissolved compounds in the membrane pores (Yigit et al. 2010).

7.1.2 Pneumatic Cleaning

Pneumatic cleaning involves the use of air under pressure for cleaning membrane, usually by airlifting, air sparging, and air scouring. The air causing a shear force destabilizes and removes foulants material from the membrane surface. The method is advantageous because it does not involve the use of chemicals, however, the use of pumping air is expensive to comply with (An et al. 2010).

7.1.3 Ultrasonic Cleaning

In ultrasonic cleaning, ultrasound waves are utilized to generate disturbance in the liquid medium. The generation of vapor bubbles and their exploding as a result of cavitation transfer energy to the membrane periphery in agitation form, consequently eliminating the accumulated materials from the surface of the membrane material. The procedure relies on numerous aspects that include ultrasonic power, temperature, crossflow velocity, and pulse duration. Ultrasonic waves are functional at a molecular level and are very effective in cleaning membranes (Kyllönen et al. 2005; Wan et al. 2013).

7.1.4 Sponge Ball Cleaning

In sponge ball cleaning, sponge balls are inserted into the reactor, and it moves through the permeate, causing the foulants to dislodge off the membrane surface. Sponge balls are prepared of materials like polyurethane and are used in membrane module units having large diameters such as tubular membrane modules (Obotey Ezugbe and Rathilal 2020).

7.2 Chemical Cleaning

Chemical cleaning techniques are utilized for irreversible membrane fouling, which is impossible to remove with physical methods. The successful application of chemical methods involves the selection of cleaning chemicals and understanding the interactions among the membrane material, foulant and cleaning chemicals (Liu et al. 2001). Chemical cleaning has a few key characteristics: (1) loose and dislodge the foulant off the membrane material; (2) retain the foulant in solution; (3) should not cause any new type of fouling; (4) and also should not deteriorate the material of the membrane.

Chemical cleaning is a cleaning in place (CIP) strategy wherein cleaning chemical is charged in the retentate channel where cleaning chemical break the bonds of the foulant material which are removed normally by the crossflow. Different chemicals are utilized for cleaning irreversible fouling, which can be classified as acids, basis/alkalis, surfactants, enzymes, chelating agents and disinfectants, etc., which are aimed at removing different kinds of fouling. For instance, various acids like phosphoric acid (H_3PO_4), hydrochloric acid (HCl) and nitric acid (HNO_3), etc. (Lin et al. 2010) are used for removal of inorganic fouling while bases/alkalis such as sodium hydroxide (NaOH), carbonates, and phosphates which are normally operated at pH 11–12 or less are employed for removing organic fouling (Obotey Ezugbe and Rathilal 2020).

7.3 Biological /biochemical Cleaning

Biological cleaning employs active biological agents like enzymes (both in single or mixture form) for membrane cleaning. Contrary to other methods, which require intensive physical and chemical environments and larger footprint areas, biological methods are sustainable and require a low footprint membrane area. Mostly applied cleaning techniques are energy uncoupling, enzymatic cleaning, and quorum quenching in membrane bioreactors for the cleaning of membranes which are employed in the cleaning of wastewater from abattoir (Maartens et al. 1996).

7.4 Physico-Chemical Cleaning Methods

In physicochemical approaches, exclusion of foulants from the membrane can be done collectively by chemical and physical methods. The combined approach increases the overall efficiency of the process, which otherwise would have been less. This includes the addition of a certain chemical to the physical process. The most typical example of the physicochemical method is chemically enhanced backwashing. Other examples include chemical cleaning assisted by ultrasonic waves, which could increase flux recoveries up to 95% (Obotey Ezugbe and Rathilal 2020; Maskooki et al. 2010).

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